

学校编码: 10384
学号: 19820100154011

分类号 _____ 密级 _____
UDC _____

厦 门 大 学

博 士 学 位 论 文

二维 Au 晶格构筑及其性质研究

Fabrication and characterization of two-dimensional Au lattices

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论文提交日期: 2015 年 05 月

论文答辩时间: 2015 年 05 月

学位授予日期: 2015 年 月

答辩委员会主席: _____

评 阅 人: _____

2015 年 06 月

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摘要

二维结构材料因其独特的对称性和缺失的第三维度具有与体材料截然不同的物理性质，包括独特的能带结构、优异的电学性能以及拓扑保护的表面态等；其固有的柔韧性、坚固性以及超薄等特点，还使得二维结构材料拥有良好的机械属性。这些新奇特性使得二维结构材料在信息领域具有广阔的应用前景，成为当今最热门的研究课题之一。而如何在广泛应用于半导体器件的硅基结构上制备稳定有序的二维材料进而实现器件上的开发和应用，成为人们日益关注的科学问题。因此，本论文着重针对硅基二维金属晶格的制备、结构表征以及性质探索等几个方面展开系统研究，具体包括以下三个部分：

1. 研究Si(111)-7×7表面上Au二维晶格的可控制备和原子构型。实验通过室温沉积并进行适当的快速退火处理，有效地控制Au在Si基底表面的排布和外延结构的形成。随着覆盖度的增加，Au-Si表面结构经历了Au₆Si₃团簇—1L Au/Si团簇层—二维Au晶格—第三层Au岛的演化过程，而Au与Si衬底间的相互作用也逐渐减弱。精确控制生长条件可制备得到大面积规则有序的二维Au晶格。对RHEED衍射图样的分析表明，对比干净的Si(111)-7×7表面，Au₆Si₃团簇出现了表面幻数团簇的衍射特征，而1L Au/Si团簇层呈现出微弱的衍射新条纹，并在二维Au晶格中形成特征的衍射条纹。通过比对Si(111)-1×1表面衍射条纹间距可知，这些特征衍射条纹来自间距约为4.6 Å的Au原子。结合覆盖度和第一性原理计算可知，相比于Au₆Si₃团簇，1L Au/Si团簇将在每个半单胞中总能最低的3个H₃位吸附Au原子以饱和7×7表面剩余的悬挂键。而在1L Au/Si团簇层之上，Au原子以密堆积的方式排布形成了由限制于半单胞内的全同Au团簇周期排列而成的二维Au晶格。

2. 利用扫描隧道显微镜/谱研究二维Au晶格的电子结构和输运特性。对比+3.0 V和+2.5 V偏压下的STM图像发现，二维Au晶格内存在很强的量子耦合效应，出现了类似于Kagome晶格的STM图像；而在低偏压和负偏压区域则表现为有序度很低的STM图像。利用扫描隧道谱表征发现，二维Au晶格的STS谱中出现了反常的非对称宽导电带隙，其带隙从-3.0 V到+1.1 V。对其起源与机制探讨表明，隧穿的电子以de Broglie波的形式在二维Au晶格中传播，在特定的电压（能量）范围内无法

向外输运，传播受到阻塞，形成带隙，而电子与空穴有效质量的不同造成了带隙的非对称性。有限元模拟显示载流子在二维Au晶格中传播产生了-3.0~+1.3 V的传播带隙，与STS谱测量的结果符合得很好，并且载流子的输运行为进一步地由二维Au晶格的结构对称性所调制。

3. 针对Au和Zn二维晶格的结构和输运性质进行了对比分析。结果表明，在结构上，两者都呈现出六角蜂窝状结构；在输运性质上，二维Zn晶格相对1L Zn/Si团簇也出现了反常增大的导电带隙，但其增大幅度明显小于Au/Si体系。对于二者的差异我们主要从三个方面来分析：（1）二维Zn晶格与二维Au晶格的原子结构不同；（2）两种晶格与Si衬底的相互作用强度不同；（3）两种二维晶格面内的耦合强度不同。对于二维Zn晶格，不同偏压下的STM图像始终是典型的六角蜂窝状结构；而二维Au晶格在不同扫描偏压下所得的STM图像存在较大差别，特别在+2.5 V偏压时STM图像中出现了量子态耦合的Kagome晶格，可见二维Au晶格面内的耦合强度要比二维Zn晶格强。而二维Au晶格与Si衬底的相互作用较弱，使得其载流子的输运行为更局限于二维晶格面内，从而形成较大的传输带隙。研究结果进一步阐明了二维金属晶格独特输运性质的物理起源和本质。

关键词：Si(111)-7×7；二维 Au 晶格；扫描隧道显微镜；反射式高能电子衍射；量子耦合；载流子输运性质

Abstract

The nature of two-dimensional (2D) materials, such as their unique symmetry and the reduced dimensionality, leads to the appearance of phenomena that are very different from their bulk materials, such as the novel band structures, electronic performance and topologically protected surface states. These materials also play an entirely mechanical role as they are inherently flexible, strong, and extremely thin. Due to their novel properties, 2D materials have promising application prospects in the field of information, and become one of the popular research areas nowadays. Furthermore, because of the most widely used Si-based materials, the preparation of 2D materials on Si-based structures to realize the development and application of the devices become an increasingly concerned topic. Thus, this thesis mainly focuses on the fabrication, structure characterization, and properties exploration of two-dimensional Au lattices on Si(111)- 7×7 surface. The major results are as follows:

1. The controllable fabrication and atomic configurations of the ordered Au low-dimensional structures on Si(111)- 7×7 surface have been studied firstly. By deposition at room temperature and rapid thermal annealing, the arrangement of Au on Si substrate could be controlled effectively. With the increase of Au coverage, the evolution of the Au-Si surface structure undergoes the transformation from Au_6Si_3 cluster- 1L Au/Si cluster-the first Au/Si layer- 2D Au lattices- Au islands. The well-ordered 2D Au lattices have been fabricated at the coverage of 0.8 ML. In the analysis of RHEED pattern, the Au_6Si_3 cluster appears the diffraction characterization of magic clusters compare to clean Si(111)- 7×7 surface, and two new faint diffraction streaks appear in the RHEED pattern of 1L Au/Si layer, which form characteristic diffraction streaks in 2D Au lattices. These characteristic diffraction streaks come from Au atoms with distance about 4.6 Å, in compare with the distance between streaks of Si(111)- 1×1 surface. Combined with the estimation of Au coverage and first-principles calculations, we speculate that for 1L Au/Si cluster about three additional Au atoms are

further adsorbed in each HUC on the H_3 site with lowest total energy, which would further saturate the remaining dangling bonds of the 7×7 surface. Subsequently, Au atoms are closely packed on the top of 1L Au/Si layer and form 2D Au lattices with identical Au clusters restricted in HUCs. The 2D Au lattices with close-packed form display good thermal stability.

2. The electronic structure and transport properties of 2D Au lattices have been investigated by STM and STS. The comparison of STM images recorded at +3.0 V and +2.5 V shows that there exist strong quantum coupling effects in 2D Au lattices. The STM image appears Kagome-like lattices at the bias of +2.5 V; while at low and negative bias voltages, the STM images turn to low degree of order. The scanning tunneling spectra of 2D Au lattices show an abnormal asymmetric gap from -3.0 V to +1.1 V. The exploration of this wide conductive gap suggests that the tunneling electrons transport in the 2D Au lattices as de Broglie wave, which might be blocked at specific voltage (energy) range and generates a transport gap. And the asymmetry of the conductive gap would be caused by the different effective mass of electrons and holes. Further finite-difference time-domain calculation shows that the electrons transport in 2D Au lattices generates a transport gap from -3.0 V to +1.3 V, which agrees well to our STS measurement. Additionally, the carrier transport behavior is further modulated by the structure of 2D Au lattices.

3. The comparative analysis on the structure and the transport properties of 2D lattices has been further performed for Au and Zn. Both 2D lattices show hexagonal honeycomb structures. An abnormal increased conductive gap was also observed in 2D Zn lattices compare to 1L Zn/Si cluster, but to a less extent than that of Au. This difference indicates the distinction in the transport behaviors between Au and Zn 2D lattices, which were mainly analyzed from three aspects: firstly, the difference of atomic structure between 2D Zn lattices and 2D Au lattices; secondly, the difference of the interaction between Si substrate and the two kinds of 2D lattices; thirdly, the difference in the strength of in-plane coupling in these two lattices. For 2D Zn lattices, the STM images always present hexagonal honeycomb structures at different bias voltages. While

the bias-dependent STM images of 2D Au lattices displays great difference. Especially at bias voltage of +2.5 V, the STM image exhibits a Kagome-like morphology due to quantum coupling effect, which suggests that there exist stronger coupling effect in 2D Au lattices. Moreover, the weak interaction of Au and Si substrate causes that the carries are even more limited to the in-plane transport for 2D Au lattices, which leads to the larger conductive gap. The results further indicate the origin of the unique carrier transport properties of the 2D metal lattices.

Keywords: Si(111)-7×7 Surface; Two-dimensional Au Lattices; STM; RHEED; Quantum coupling; Carrier transport properties.

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第一章 绪论

随着信息科学与技术的飞速发展，传统器件尺度日益减小，基于描述电子和光子等在三维体材料中行为的经典物理学的局限性越来越突出，人们逐渐采用以量子力学和相对论为基础的近代物理学描述粒子在二维、一维及零维等结构中的行为。二维结构材料具有丰富的新奇特性，在信息领域具有广阔的应用前景，因此成为当今最热门的研究课题之一。

1.1 二维结构材料的分类

二维结构是指在二维平面内原子及成键结构相似，而在垂直方向上成键强度要远弱于平面的一类材料结构。由于电荷输运和热输运被限制在二维平面内而导致不同寻常的物理性质，二维结构材料自上世纪 60 年代起就广受人们的关注，例如，1966 年，Frindt 和 Joensen 等就探讨了几个分子层厚度单晶 MoS_2 的制备及性质^[1,2]；1969 年，Wilson 等报导了过渡金属硫化物的制备及光学、电学与结构性质^[3]；1971 年，Consador 等报导了利用超导层状结构单晶 NbSe_2 制备的器件等^[4]。在 2004 年由 Novoselov 和 Geim 发现了单层石墨烯^[5]，激发了人们对包括石墨烯在内的二维结构材料的研究热情。

到目前为止，已经被制备和研究的二维材料多达几十种，这些二维结构材料主要可以分为以下三大类，如表 1.1 所示^[6]。

第一类可以称之为石墨烯家族，包括：石墨烯^[7-13]、六方氮化硼 ($h\text{-BN}$)^[14-18]、硅烯^[19-22]、锗烯^[23-25]、杂化硼碳氮 (BCN)^[26-28]、氟化石墨烯^[29,30]、氢化石墨烯 (石墨烷)^[31]、氧化石墨烯等^[32]。这其中石墨烯无疑是整个二维材料体系中最受瞩目的一种，其良好的机械强度以及晶体和电子性质使其具有广阔的应用前景。石墨烯家族在结构上与石墨烯类似，都是蜂窝状有序排列的晶格结构 (如图 1.1 (a) 所示)，多具有良好的润滑性、导热性及耐化学腐蚀性等机械属性，但在电学等性质上也存在较大的差异。例如：组成 $h\text{-BN}$ 的氮原子和硼原子与石墨烯的碳原子均以 sp^2 轨道杂化的形式形成共价键，键角成 120° ， $h\text{-BN}$ 可作为石墨烯良好的衬底，

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